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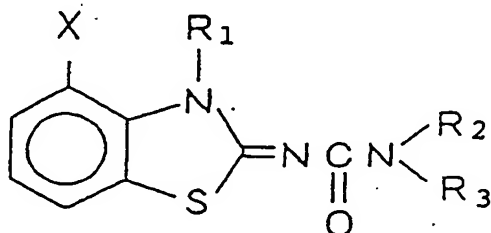
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(54) Benzothiazole derivative and agricultural and horticultural fungicide composition containing the same.

(57) A benzothiazole derivative represented by formula (I):



(I)

wherein R<sub>1</sub> represents a lower alkyl, alkenyl, alkynyl, cycloalkyl or alkoxyalkyl group, a benzyl group or an alkyl or alkoxyalkyl group containing a carbonyl group; R<sub>2</sub> represents a hydrogen atom or a lower alkyl group; R<sub>3</sub> represents an alkyl group having from 1 to 3 carbon atoms and containing from 1 to 6 fluorine atoms or a group represented by -O-R<sub>4</sub> wherein R<sub>4</sub> represents a hydrogen atom, a lower alkyl group or a benzyl group; and X represents a halogen atom or a hydrogen atom, and a fungicide composition comprising the same are disclosed.

1,1-diethylmethyl, 1-methylbutyl, t-pentyl, 1,2-dimethylpropyl, cyclobutylmethyl, cyclopropylethyl, 2-methyl-cyclopropylmethyl, n-hexyl, i-hexyl, 1-methylpentyl, 1,3-dimethylbutyl, 1,2,2-trimethylpropyl, 1-ethylbutyl, 1-ethyl-2-methylpropyl, cyclopentylmethyl, cyclobutylethyl, cyclopropylpropyl, 1,1,2-trimethylpropyl, 1,1-dimethylbutyl, 2,2-dimethylbutyl, 2-ethylbutyl, 2-methylpentyl, 2,3-dimethylbutyl, 3-methylpentyl, 3,3-dimethylbutyl, n-heptyl, 5-methylhexyl, 1-methylhexyl, 1,4-dimethylpentyl, 1,3-dimethylpentyl, 1,3,3-trimethylbutyl, 1-methyl-2-ethylbutyl, 2-methylhexyl, 2,4-dimethylpentyl, 2,3-dimethylpentyl, 1,1-dimethylpentyl, 2,2-dimethylpentyl, 3,3-dimethylpentyl, 4,4-dimethylpentyl, 3-methylhexyl, 3,4-dimethylpentyl, 4-methylhexyl, cyclohexylmethyl, cyclopentylethyl, cyclobutylpropyl, cyclopropylbutyl, n-octyl, 6-methylheptyl, 1-methylheptyl, 1,1-dimethylhexyl, 2-methylheptyl, 3-methylheptyl, 4-methylheptyl, 5-methylheptyl, 2,2-dimethylhexyl, 3,3-dimethylhexyl, 4,4-dimethylhexyl, 5,5-dimethylhexyl, cyclohexylethyl, cyclopentylpropyl, cyclobutylbutyl and cyclopropylpentyl groups. Among them, methyl, ethyl, n-propyl, i-propyl, n-butyl, cyclopropylmethyl, n-hexyl and i-butyl groups are preferred.

The lower alkenyl group represented by  $R_1$  in formula (I) preferably contains from 2 to 6 carbon atoms, more preferably from 3 to 4 carbon atoms. Specific examples thereof include vinyl, 1-propenyl, 2-propenyl, i-propenyl, 1-butenyl, 2-butenyl, 3-butenyl, 2-methyl-1-propenyl, 2-methyl-2-propenyl, 1-pentenyl, 3-pentenyl, 4-pentenyl, 1-methyl-3-butenyl, 1-methyl-2-butenyl, 1,2-dimethyl-2-propenyl, 1,1-dimethyl-2-propenyl, 1-hexenyl, 3-hexenyl, 4-hexenyl, 5-hexenyl, 1-methyl-2-pentenyl, 1-methyl-3-pentenyl, 1-methyl-4-pentenyl, 1,4-dimethyl-2-butenyl, 1,1-dimethyl-2-butenyl and 1,1-dimethyl-3-butenyl groups. Among them, 2-propenyl, 2-butenyl and 2-methyl-2-propenyl groups are preferred.

The lower alkynyl group represented by  $R_1$  in formula (I) preferably contains from 2 to 6 carbon atoms, more preferably 3 carbon atoms. Specific examples thereof include ethynyl, 2-propynyl, 1-propynyl, 1-butylnyl, 2-butylnyl, 3-butylnyl, 1-methyl-2-propynyl, 1-pentylnyl, 2-pentylnyl, 3-pentylnyl, 4-pentylnyl, 1-methyl-3-butylnyl, 1-methyl-2-butylnyl, 1,1-dimethyl-2-propynyl, 1-hexynyl, 2-hexynyl, 3-hexynyl, 4-hexynyl, 5-hexynyl, 1-methyl-2-pentylnyl, 1-methyl-3-pentylnyl, 1-methyl-4-hexynyl, 1,1-dimethyl-2-butylnyl and 1,1-dimethyl-3-butylnyl groups. Among them, 2-propynyl group is preferred.

The lower cycloalkyl group represented by  $R_1$  in formula (I) preferably contains from 3 to 6 carbon atoms, more preferably 5 carbon atoms. Specific examples thereof include cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl groups. Among them, cyclopentyl group is preferred.

The alkoxyalkyl group represented by  $R_1$  in formula (I) preferably contains from 2 to 5 carbon atoms, more preferably from 2 to 4 carbon atoms. Specific examples thereof include methoxymethyl, methoxyethyl, ethoxymethyl, propoxymethyl, ethoxyethyl, methoxypropyl, i-propoxymethyl, 1-methoxy-2-propyl, 2-methoxy-2-propyl, butoxymethyl, propoxyethyl, ethoxypropyl, methoxybutyl, i-butoxymethyl, t-butoxymethyl, sec-butoxymethyl, i-propoxyethyl and 2-ethoxy-1-methylethyl groups.

The alkyl or alkoxyalkyl group containing a carbonyl group represented by  $R_1$  in formula (I) preferably has one carbonyl group and contains from 2 to 5 carbon atoms, more preferably from 2 to 4 carbon atoms, in total. Specific examples thereof include carboxymethyl, acetylmethyl, methoxycarbonylmethyl, acetylethyl, propionylmethyl, methoxycarbonylethyl, ethoxycarbonylmethyl, carboxypropyl, acetylpropyl, propionylethyl, butyrylmethyl, i-butyrylmethyl, methoxycarbonylpropyl, propoxycarbonylmethyl, ethoxycarbonylethyl, carboxybutyl, 1-methoxycarbonyl-2-propyl groups.

Among the alkoxyalkyl groups and the alkyl or alkoxyalkyl groups containing a carbonyl group, carboxymethyl, methoxymethyl, methoxyethyl, acetylmethyl and ethoxycarbonylmethyl groups are preferred.

The lower alkyl group represented by  $R_2$  in formula (I) preferably contains from 1 to 3 carbon atoms, more preferably from 1 carbon atom. Specific examples thereof include methyl, ethyl, n-propyl and i-propyl groups. Among the groups represented by  $R_2$ , hydrogen atom and methyl group are preferred.

Specific examples of the  $C_{1-3}$  alkyl group containing 1 to 6 fluorine atom represented by  $R_3$  in formula (I) include trifluoromethyl, 2,2,2-trifluoroethyl, 2,2-difluoroethyl, 2-fluoroethyl, 1,2,2,2-tetrafluoroethyl, 1,1,2,2,2-pentafluoroethyl, 3,3,3-trifluoropropyl, 2,2,3,3,3-pentafluoropropyl, 2,3,3,3-tetrafluoropropyl, 3-fluoropropyl, 3,3-difluoropropyl, 1,1,2,2,3,3-hexafluoropropyl, 1,2,2,3,3,3-hexafluoropropyl and 1,1,2,3,3,3-hexafluoropropyl groups. Among them, 2,2,2-trifluoroethyl, 2,2,3,3,3-pentafluoropropyl and 2-fluoroethyl groups are preferred.

The lower alkyl group represented by  $R_4$  in formula (I) preferably contains from 1 to 8 carbon atoms, more preferably from 1 to 7 carbon atoms. Specific examples thereof include methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl, sec-butyl, cyclopropylmethyl, n-pentyl, i-pentyl, 2-methylbutyl, neopentyl, 1,1-diethylmethyl, 1-methylbutyl, t-pentyl, 1,2-dimethylpropyl, cyclobutylmethyl, cyclopropylethyl, 2-methyl-cyclopropylmethyl, n-hexyl, i-hexyl, 1-methylpentyl, 1,3-dimethylbutyl, 1,2,2-trimethylpropyl, 1-ethylbutyl, 1-ethyl-2-methylpropyl, cyclopentylmethyl, cyclobutylethyl, cyclopropylpropyl, 1,1,2-trimethylpropyl, 1,1-dimethylbutyl, 2,2-dimethylbutyl, 2-ethylbutyl, 2-methylpentyl, 2,3-dimethylbutyl, 3-methylpentyl, 3,3-

If desired, the compound may be used as admixture with or in combination with other agricultural chemicals such as herbicides, insecticides and other fungicides, or fertilizers.

The compound of the present invention should be applied in such an amount sufficient for obtaining desired effects. For example, the compound of formula (I) wherein  $R_3$  is  $-O-R_4$  may be applied in an amount of from 20 to 2,000 g/ha, and the compound of formula (I) wherein  $R_3$  is a  $C_{1-3}$  alkyl group containing 1 to 6 fluorine atoms may be applied in an amount of from 50 to 2,000 g/ha. In general, a suitable amount of the compound to be applied ranges from 100 to 1,000 g/ha, which is applied in the form of a wettable powder, an emulsion, a dust, a granule, or a suspension containing the active ingredient in a concentration of from 0.1 to 50 % by weight.

Emulsions can be prepared by dissolving the active ingredient in an agriculturally acceptable organic solvent and adding a solvent-soluble emulsifying agent. Suitable solvents include xylene, o-chlorotoluene, cyclohexanone, isophorone, dimethylformamide, dimethyl sulfoxide, and a mixture thereof. An aromatic hydrocarbon or a mixture of an aromatic hydrocarbon, a ketone and a polar solvent is particularly preferred. A surfactant as an emulsifying agent is added in an amount of from 1 to 20% by weight based on the emulsion. Any of anionic, cationic or non-ionic surfactants may be employed. A suitable concentration of the active ingredient in the emulsion is from 0.5 to 50% by weight, preferably from 5 to 30% by weight, as for the compound of formula (I) wherein  $R_3$  is  $-O-R_4$ ; and from 0.5 to 20% by weight, preferably from 1 to 10% by weight, as for the compound of formula (I) wherein  $R_3$  is a  $C_{1-3}$  alkyl group containing 1 to 6 fluorine atoms.

Examples of suitable anionic surfactants include alkylsulfuric esters, alkyl diphenyl ether disulfonates, naphthylmethanesulfonates, lignin sulfonates, alkylsulfosuccinates, alkylbenzene sulfonates, and alkylphosphates. Examples of suitable cationic surfactants include alkylamine salts and quaternary ammonium salts. Examples of suitable nonionic surfactants include polyoxyethylene alkyl ethers, polyoxyethylene alkylaryl ethers, sorbitan fatty acid esters, polyoxyethylene sorbitan fatty acid esters, polyoxyethylene sorbitol fatty acid esters, glycerin fatty acid esters, and polyoxyethylene fatty acid esters.

Wettable powders can be prepared by adding the active ingredient to a finely pulverized inert solid carrier and a surfactant. Wettable powders generally contain from 2 to 50 % by weight of the active ingredient and from 1 to 20 % by weight of the surfactant. Suitable inert finely pulverized solid carriers include naturally-occurring clay, silicates, silica, and alkaline earth metal carbonates. Typical examples of such solid carriers are kaolin, zeolite, talc, diatomaceous earth, magnesium carbonate, calcium carbonate, and dolomite. Commonly employed anionic or nonionic surfactants (specific examples thereof have been described above) or mixtures thereof can be used as an emulsifying agent, a spreading agent or a dispersing agent.

Dusts can be prepared by adding the active ingredient to a commonly employed inert carrier, such as talc, a fine powder of clay, pyrophyllite, diatomaceous earth or magnesium carbonate, in a concentration ranging from 0.1 to 20% by weight, preferably from 0.5 to 5 % by weight.

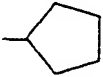

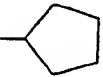
Granules can be prepared by mixing the active ingredient with a finely pulverized inert carrier, such as bentonite, diatomaceous earth, kaolin, clay or talc, kneading the obtained mixture with water, and granulating by means of a granulating apparatus. Alternatively, granules may be prepared by adhering a solution of the active ingredient and a spreading agent onto a carrier having previously been granulated to a particle size of from about 15 to 30 mesh or a granular mineral, such as naturally-occurring pumice powder, acid clay or zeolite, having its particle size previously been regulated by grinding. Granules suitably contain the active ingredient in a concentration ranging from 0.2 to 20 % by weight, preferably from 1 to 10 % by weight.

Flowables can be prepared by finely pulverizing the active ingredient and mixing the resulting powder with a surfactants and water. Any of the above-mentioned anionic, cationic or nonionic surfactants can be used either individually or in combination thereof in an amount of from 1 to 20 % by weight. The content of the active ingredient in the flowable ranges from 1 to 50 % by weight, and preferably from 2 to 20 % by weight.

The compound of formula (I) is useful as an active ingredient of agricultural or horticultural fungicidal compositions and exhibits excellent controlling effects on rice blast, apple scab, pear scab, grape downy mildew, blue mold on various crops, powdery mildew or rust on various crops, and wheat leaf spot. The compound of the invention produces not only preventive effects but excellent curing effects and is therefore applicable for disease control after infection. The compound exhibits asmotranslational activity and is therefore applicable to soil for cauline and foliar disease control.

Typical examples of the compounds according to the present invention are shown in Table 1.

TABLE 1 (cont'd)

Compound No.	$-R_1$	$-R_2$	$-R_3$	$-X$	m.p. (°C)
20		-H	-O-CH <sub>3</sub>	-H	45-48
21	-CH <sub>2</sub> (CH <sub>2</sub> ) <sub>4</sub> CH <sub>3</sub>	-H	-O-CH <sub>3</sub>	-H	66-68
22	-CH <sub>2</sub> -C <sub>6</sub> H <sub>5</sub>	-H	-O-CH <sub>3</sub>	-H	106-107
23	-CH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	-H	-O-CH <sub>3</sub>	-H	129-130
24	-CH <sub>2</sub> CH <sub>2</sub> OCH <sub>3</sub>	-H	-O-CH <sub>3</sub>	-H	124-125
25	-CH <sub>2</sub> OCH <sub>3</sub>	-H	-O-CH <sub>3</sub>	-H	116-118
26	-CH <sub>2</sub> CH=CH <sub>2</sub>	-H	-O-CH <sub>3</sub>	-F	
27	-CH <sub>2</sub> CH=CH <sub>2</sub>	-H	-O-CH <sub>3</sub>	-Cl	
28	-CH <sub>2</sub> - 	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	141- 142.5
29	-CH <sub>2</sub> -C <sub>6</sub> H <sub>5</sub>	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	182-183
30	-CH <sub>2</sub> CH-CH <sub>3</sub>   CH <sub>3</sub>	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	140-141
31		-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	165-166
32	-CH <sub>2</sub> (CH <sub>2</sub> ) <sub>4</sub> CH <sub>3</sub>	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	117-118
33	-CH <sub>2</sub> (CH <sub>2</sub> ) <sub>2</sub> CH <sub>3</sub>	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	110-111
34	-CH <sub>2</sub> CH <sub>2</sub> CH <sub>3</sub>	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	135- 136.5
35	-CH <sub>2</sub> C=CH <sub>2</sub>   CH <sub>3</sub>	-H	-CH <sub>2</sub> CF <sub>3</sub>	-H	125-126

SYNTHESIS EXAMPLE 2

## Synthesis of 3-Allyl-2-N-methoxycarbamoyliminobenzothiazole (Compound No. 6)

5 In 100 ml of methylene chloride was dissolved 3.0 g of trichloromethyl chloroformate, and the solution was cooled to 0 °C. To the solution was added dropwise a solution of 5.7 g of 3-allyl-2-iminobenzothiazole (the compound of formula (II) wherein -R<sub>1</sub> is -CH<sub>2</sub>CH=CH<sub>2</sub>) and 2.4 g of pyridine in 50 ml of methylene chloride at 0 to 5 °C, followed by stirring for 2 hours. To the mixture was added 4.2 g of O-methylhydroxylamine (the compound of formula (IV) wherein -R<sub>2</sub> is -H and -R<sub>3</sub> is -O-CH<sub>3</sub>), followed by stirring at the same temperature for 4 hours. The reaction mixture was washed successively with diluted hydrochloric acid, a sodium hydrogencarbonate aqueous solution, water, and a saturated sodium chloride aqueous solution, dried over anhydrous magnesium sulfate, and distilled under reduced pressure to remove the solvent. The crude product thus obtained was purified by silica gel column chromatography to obtain 4.8 g (yield: 61 %) of the titled compound. Melting Point: 110-111 °C.

SYNTHESIS EXAMPLE 3

## Synthesis of 3-Allyl-2-N-methyl-N-methoxycarbamoyliminobenzothiazole (Compound No. 7)

20 In 100 ml of methylene chloride was dissolved 3.0 g of trichloromethyl chloroformate, and the solution was cooled to 0 °C. To the solution was added dropwise a solution of 5.7 g of 3-allyl-2-iminobenzothiazole (the compound of formula (II) wherein -R<sub>1</sub> is -CH<sub>2</sub>CH=CH<sub>2</sub>) and 2.4 g of pyridine in 50 ml of methylene chloride at 0 to 5 °C, followed by stirring for 2 hours. To the mixture was added 4.2 g of N,O-dimethylhydroxylamine (the compound of formula (IV) wherein -R<sub>2</sub> is -CH<sub>3</sub> and -R<sub>3</sub> is -CH<sub>3</sub>), followed by stirring at the same temperature for 4 hours. The reaction mixture was washed successively with diluted hydrochloric acid, a sodium hydrogencarbonate aqueous solution, water, and a saturated sodium chloride aqueous solution, dried over anhydrous magnesium sulfate, and distilled under reduced pressure to remove the solvent. The crude product thus obtained was purified by silica gel column chromatography to obtain 4.8 g (yield: 58 %) of the titled compound. Melting Point: 62-63 °C.

SYNTHESIS EXAMPLE 4

## Synthesis of 3-Allyl-2-N-n-propoxycarbamoyliminobenzothiazole (Compound No. 9)

35 In 100 ml of methylene chloride was dissolved 3.0 g of trichloromethyl chloroformate, and the solution was cooled to 0 °C. To the solution was added dropwise a solution of 5.7 g of 3-allyl-2-iminobenzothiazole (the compound of formula (II) wherein -R<sub>1</sub> is -CH<sub>2</sub>CH=CH<sub>2</sub>) and 2.4 g of pyridine in 50 ml of methylene chloride at 0 to 5 °C, followed by stirring for 2 hours. To the mixture was added 6.8 g of O-n-propylhydroxylamine (the compound of formula (IV) wherein -R<sub>2</sub> is -H and -R<sub>3</sub> is -CH<sub>2</sub>CH<sub>2</sub>CH<sub>3</sub>), followed by stirring at the same temperature for 4 hours. The reaction mixture was washed successively with diluted hydrochloric acid, a sodium hydrogencarbonate aqueous solution, water, and a saturated sodium chloride aqueous solution, dried over anhydrous magnesium sulfate, and distilled under reduced pressure to remove the solvent. The crude product thus obtained was purified by silica gel column chromatography to obtain 4.7 g (yield: 57 %) of the titled compound. Melting Point: 80-81 °C.

SYNTHESIS EXAMPLE 5

## Synthesis of 3-Allyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole (Compound No. 38)

50 In 100 ml of methylene chloride was dissolved 3.0 g of trichloromethyl chloroformate, and the solution was cooled to 0 °C. To the solution was added dropwise a solution of 5.7 g of 3-allyl-2-iminobenzothiazole and 2.4 g of pyridine in 50 ml of methylene chloride at 0 to 5 °C, followed by stirring for 2 hours. To the mixture was added 8.9 g of 2,2,2-trifluoroethylamine, followed by stirring at the same temperature for 4 hours. The reaction mixture was washed successively with diluted hydrochloric acid, a sodium hydrogencarbonate aqueous solution, water, and a saturated sodium chloride aqueous solution, dried over anhydrous magnesium sulfate, and distilled under reduced pressure to remove the solvent. The crude product thus obtained was purified by silica gel column chromatography to obtain 5.6 g (yield: 59 %) of the titled compound. Melting Point: 132.5-134.5 °C.

FORMULATION EXAMPLE 3 (Dust)	
Component	Amount (part by weight)
Compound No. 10 Kaolin clay	4 96

The above components were mixed and ground to obtain a dust according to the present invention.

FORMULATION EXAMPLE 4 (Granule)	
Component	Amount (part by weight)
Compound No. 21	5
Bentonite	45
Talc	45
Sodium lignin sulfonate	5

The above components were uniformly mixed and ground, kneaded together with water, granulated, and dried to obtain a granule according to the present invention.

FORMULATION EXAMPLE 5 (Flowable)	
Component	Amount (part by weight)
Compound No. 15	10
Ethylene glycol	5
Sorpol 3078	5
Sorpol 7512	1
Water	79
(Sorpol 3078: a trademark of a product comprising a polyoxyethylene alkylphenyl ether sulfate; Sorpol 7512: a trademark of a product; each manufactured by Toho Chemical Industry Co., Ltd.)	

The above components were uniformly mixed and ground to prepare a flowable according to the present invention.

FORMULATION EXAMPLE 6 (Emulsion)	
Component	Amount (part by weight)
Compound No. 8	10
Isophorone	23
o-Chlorotoluene	28
Xylene	23
Sorpol 900A	8
Sorpol 900B	8
(Sorpol 900 A: a trademark of a product comprising a polyoxyethylenealkylphenol polymer, a polyoxyethylene alkylaryl ether, a polyoxyethylene sorbitan fatty acid ester and an anionic surfactant; manufactured by Toho Chemical Industry Co., Ltd.)	

The above components were uniformly mixed and dissolved to obtain an emulsion according to the present invention.

The above components were uniformly mixed and ground to prepare a flowable according to the present invention.

FORMULATION EXAMPLE 12 (Emulsion)	
Component	Amount (part by weight)
Compound No. 30	10
Isophorone	23
o-Chlorotoluene	28
Xylene	23
Sorpol 900A	8
Sorpol 900B	8

The above components were uniformly mixed and dissolved to obtain an emulsion according to the present invention.

#### TEST EXAMPLE 1

##### Preventive Effect Against Wheat Powdery Mildew:

Wheat plants (variety: *Norin No. 61*) were cultivated in a plastic pot having a diameter of 7 cm. At the difoliate stage, 10 ml of an emulsion of a test compound shown in Table 2 below prepared in accordance with Formulation Example 1 and adjusted to a concentration of 500 ppm was sprayed over the plant using a spray gun. Regarding each test compound, four lots each having 13 plants were employed, and two of them were treated with the test compound (test lot) and the other two of them were not treated with the test compound (control lot). One day after the treatment, conidia of *Erysiphe graminis* which causes wheat powdery mildew were inoculated by spraying. The plants were cultivated at 20 °C in the dark for 12 hours followed by at 20 °C for 6 days with 12-hour photoperiod. On the 7th day after the inoculation, the lesions of the first and second leaves were examined, and the preventive value was calculated in accordance with the following Equation (1).

Equation (1):

$$\text{Prevention value} = \frac{(\text{Rate of lesion spot of control lot}) - (\text{Rate of lesion spot of test lot})}{(\text{Rate lesion spot of control lot})} \times 100$$

The results obtained are shown in Table 2.

TABLE 2 (cont'd)

	<u>Compound No.</u>	<u>Preventive Value</u>
5	25	100
	26	100
10	27	100

TEST EXAMPLE 2

## Preventive Effect Against Rice Blast:

Rice (variety: *Koshihikari*) was cultivated in a plastic pot having a diameter of 7 cm. At the difoliate stage, 10 ml of a wettable powder of a test compound shown in Table 3 below prepared in accordance with Formulation Example 2 and adjusted to a concentration of 500 ppm was sprayed over the plant using a spray gun. Regarding each test compound, four lots each having 10 plants were employed, and two of them were treated with the test compound (test lot), and the other two of them were not treated with the test compound (control lot). One day after the treatment, a conidium suspension of *Pyricularia oryzae* which causes rice blast was inoculated. The plants were cultivated under moisture-saturation at 25 °C in the dark for 48 hours followed by at 25 °C for 6 days with 12-hour photoperiod. On the 7th day after the inoculation, the lesions of the first and second leaves were examined, and a preventive value was calculated in the same manner as in Test Example 1. The results obtained are shown in Table 3.



TABLE 3 (cont'd)

	<u>Compound No.</u>	<u>Preventive Value</u>
5	25	100
	26	100
10	27	100

TEST EXAMPLE 3

15

Preventive Effect Against Wheat Powdery Mildew:

Wheat (species: *Norin No. 61*) was cultivated in a plastic pot having a diameter of 7 cm. At the difoliate stage, 10 ml of an emulsifiable concentrate of a test compound shown in Table 4 below prepared in accordance with Formulation Example 7 and adjusted to a concentration of 500 ppm was sprayed over the plant using a spray gun. Regarding each test compound, four lots each having 13 plants were employed, and two of them were treated with the test compound (test lot) and the other two of them were not treated with the test compound (control lot). One day after the treatment, conidia of *Erysiphe graminis* which causes wheat powdery mildew were inoculated by spraying. The plants were cultivated at 20°C in the dark for 12 hours followed by at 20°C for 6 days with 12-hour photoperiod. On the 7th day after the inoculation, the lesions of the first and second leaves were examined, and the preventive value was calculated in the same manner as in Test Example 1.

TABLE 4

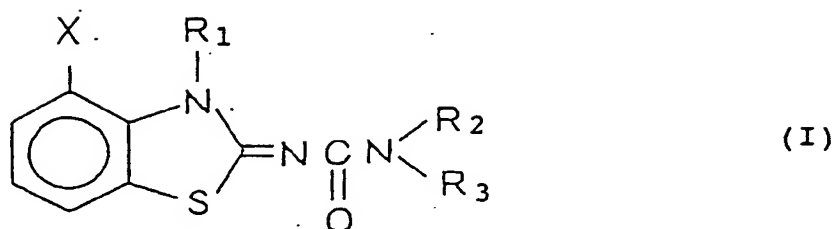
30

Compound No.	Preventive Value
28	100
29	100
30	100
31	100
32	100
33	100
34	100
35	100
36	100
37	100
38	100
39	100
40	100
41	100
42	100
43	100
44	100
45	100
46	100
47	100
48	100

45

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wherein R<sub>1</sub> represents a lower alkyl, alkenyl, alkynyl, cycloalkyl or alkoxyalkyl group, a benzyl group or an alkyl or alkoxyalkyl group containing a carbonyl group; R<sub>2</sub> represents a hydrogen atom or a lower alkyl group; R<sub>3</sub> represents an alkyl group having from 1 to 3 carbon atoms and containing from 1 to 6 fluorine atoms or a group represented by -O-R<sub>4</sub> wherein R<sub>4</sub> represents a hydrogen atom, a lower alkyl group or a benzyl group; and X represents a halogen atom or a hydrogen atom.

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2. The benzothiazole derivative of claim 1, which is selected from 3-methyl-2-N-methoxycarbamoyliminobenzothiazole, 3-ethyl-2-N-methoxycarbamoyliminobenzothiazole, 3-ethyl-2-N-ethoxycarbamoyliminobenzothiazole, 3-n-propyl-2-N-methoxycarbamoyliminobenzothiazole, 3-i-propyl-2-N-methoxycarbamoyliminobenzothiazole, 3-allyl-2-N-methoxycarbamoyliminobenzothiazole, 3-allyl-2-N-methyl-N-methoxycarbamoyliminobenzothiazole, 3-allyl-2-N-ethoxycarbamoyliminobenzothiazole, 3-allyl-2-N-n-propoxycarbamoyliminobenzothiazole, 3-allyl-2-N-(2-propoxy)carbamoyliminobenzothiazole, 3-allyl-2-N-n-butoxycarbamoyliminobenzothiazole, 3-allyl-2-N-benzyloxycarbamoyliminobenzothiazole, 3-allyl-2-N-hydroxycarbamoyliminobenzothiazole, 3-propynyl-2-N-methoxycarbamoyliminobenzothiazole, 3-n-butyl-2-N-methoxycarbamoyliminobenzothiazole, 3-i-butyl-2-N-methoxycarbamoyliminobenzothiazole, 3-cyclopropylmethyl-2-N-methoxycarbamoyliminobenzothiazole, 3-(2-methyl-2-propenyl)-2-N-methoxycarbamoyliminobenzothiazole, 3-(2-butenyl)-2-N-methoxycarbamoyliminobenzothiazole, 3-cyclopentyl-2-N-methoxycarbamoyliminobenzothiazole, 3-n-hexyl-2-N-methoxycarbamoyliminobenzothiazole, 3-benzyl-2-N-methoxycarbamoyliminobenzothiazole, 3-ethoxycarbonylmethyl-2-N-methoxycarbamoyliminobenzothiazole, 3-methoxyethyl-2-N-methoxycarbamoyliminobenzothiazole, 3-methoxymethyl-2-N-methoxycarbamoyliminobenzothiazole, 3-allyl-4-fluoro-2-N-methoxycarbamoyliminobenzothiazole, 3-allyl-4-chloro-2-N-methoxycarbamoyliminobenzothiazole, 3-cyclopropylmethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-benzyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-i-butyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-cyclopentyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-n-hexyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-n-butyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-n-propyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-(2-methyl-2-propenyl)-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-i-propyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-ethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-allyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-(2-propynyl)-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-allyl-2-N-(2,2,3,3,3-pentafluoropropyl)carbamoyliminobenzothiazole, 3-allyl-2-N-(2-fluoroethyl)carbamoyliminobenzothiazole, 3-ethoxycarbonylmethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-carboxymethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-acetylmethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-methoxyethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-methoxymethyl-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, 3-allyl-4-fluoro-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole, and 3-allyl-4-chloro-2-N-(2,2,2-trifluoroethyl)carbamoyliminobenzothiazole.
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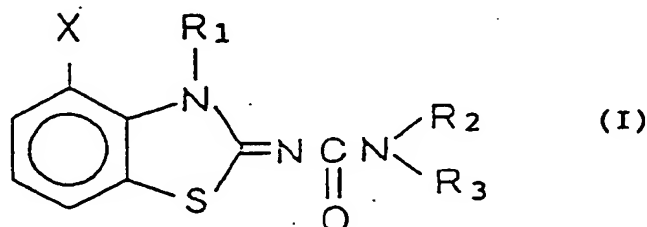
3. A fungicide composition comprising a benzothiazole derivative represented by formula (I):
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(b) The compound according to the general formula (III) is then reacted with an amine compound of the following formula (IV):



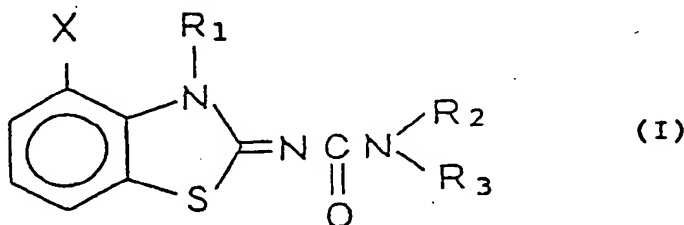
at a temperature of from 0 to 30 °C to obtain the desired compound (I).

5. A process according to claim 4 wherein in step (a) the solvent is a halogenated hydrocarbon, preferably chloroform or methylene chloride, the base is selected from pyridine, triethylamine, sodium hydrogen-carbonate or potassium hydrogencarbonate, and the reaction temperature is from 0 to 5 °C, and in step (b), the reaction temperature is from 0 to 5 °C.
6. A process for preparing a fungicide composition comprising a benzothiazole derivative represented by formula (I):



wherein R<sub>1</sub> represents a lower alkyl, alkenyl, alkynyl, cycloalkyl or alkoxyalkyl group, a benzyl group or an alkyl or alkoxyalkyl group containing a carbonyl group; R<sub>2</sub> represents a hydrogen atom or a lower alkyl group; R<sub>3</sub> represents an alkyl group having from 1 to 3 carbon atoms and containing from 1 to 6 fluorine atoms or a group represented by -O-R<sub>4</sub> wherein R<sub>4</sub> represents a hydrogen atom, a lower alkyl group or a benzyl group; and X represents a halogen atom or a hydrogen atom; wherein said derivative of formula (I) is brought into contact with a suitable carrier in order to obtain a fungicidal composition containing the active ingredient in a concentration of from 0.1 to 50 % by weight.

7. The use of a benzothiazole derivative represented by formula (I):



wherein R<sub>1</sub> represents a lower alkyl, alkenyl, alkynyl, cycloalkyl or alkoxyalkyl group, a benzyl group or an alkyl or alkoxyalkyl group containing a carbonyl group; R<sub>2</sub> represents a hydrogen atom or a lower alkyl group; R<sub>3</sub> represents an alkyl group having from 1 to 3 carbon atoms and containing from 1 to 6 fluorine atoms or a group represented by -O-R<sub>4</sub> wherein R<sub>4</sub> represents a hydrogen atom, a lower alkyl group or a benzyl group; and X represents a halogen atom or a hydrogen atom, as active ingredient in an agricultural or horticultural fungicidal composition.



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## EUROPEAN SEARCH REPORT

Application Number

EP 93 10 7070

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	EP-A-0 407 621 (HODOGAYA CHEMICAL COMPANY LIMITED) * page 19 - page 20; claims *	1,3,4,6-8	C07D277/82 A01N47/36
A	EP-A-0 337 616 (SCHERING AGROCHEMICALS LIMITED) * the whole document *	1,3,6-8	
D,A	US-A-3 671 531 (WILLIAM D. DIXON) * the whole document *	1,3	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C07D
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 08 JULY 1993	Examiner HENRY J.C.
<b>CATEGORY OF CITED DOCUMENTS</b>			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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